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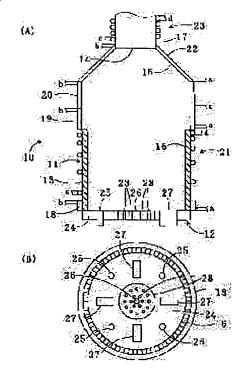
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# (54) APPARATUS FOR MANUFACTURING FULLERENE

# (57) Abstract:

PROBLEM TO BE SOLVED: To provide an apparatus for manufacturing fullerene in which fullerene is manufactured and recovered in large quantities, at a low cost and easily, where the formation and stabilization of a precursor of fullerene is secured by controlling a temperature distribution in a reaction furnace by a combustion method.

SOLUTION: The apparatus 10 for manufacturing fullerene by combusting a carbon-containing compound used as a raw material and an oxygen-containing gas in a reaction furnace 11 is equipped with a burner 12 having inlets for supplying a carbon-containing compound 25, 26 and those for supplying the oxygen-containing gas 27, 28. At the outside of the reaction



furnace 11, a plurality of outer cooling parts 18, 19, 20, 21, 22, whose cooling capacities are independently controllable, are installed in a multistage manner against an moving direction of a gas flow of a combustion product formed by a combustion of the carbon-containing compound under the oxygen-containing gas.

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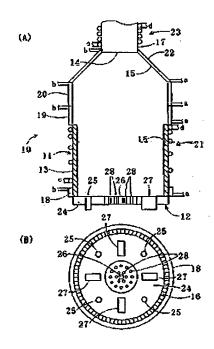
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#### (54) 【発明の名称】 フラーレンの製造装配

#### (57)【要約】

【課題】 燃焼法によるフラーレンの製造において、フラーレンの反応炉内の温度分布を制御することによりフラーレン前駆体の形成と安定化を確保してフラーレンを大量、安価、容易に製造して回収することが可能なフラーレンの製造装置を提供する。

【解決手段】 炭素含有化合物供給口25、26と酸素含有ガス供給口27、28とを有する燃焼用バーナ部12を備える反応炉11で、原料となる炭素含有化合物と酸素含有ガスとを燃焼させてフラーレンを製造するフラーレンの製造装置10であって、反応炉11の外側には、冷却能力を独立して調御可能な複数の外側冷却部18、19、20、21、22を、炭素含有化合物が酸素含有ガスの下で燃焼して形成される燃焼生成ガス流の移動方向に対して多段に設けた。



#### 【特許請求の衛囲】

【請求項1】 炭素含有化合物供給口と酸素含有ガス供給口とを有する燃焼用バーナ部を備える反応炉で、原料となる炭素含有化合物と酸素含有ガスとを燃焼させてフラーレンを製造するフラーレンの製造装置であって、前記反応炉の外側には、冷却能力を独立して制御可能な複数の外側冷却部を、前記炭素含有化合物が前記酸素含有ガスの下で燃煙して形成される燃焼生成ガス流の移動方向に対して多段に設けたことを特徴とするフラーレンの製造装置。

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【請求項2】 請求項1記載のフラーレンの製造装置に おいて、前記各外側冷却部が冷却管又は冷却ジャケット のいずれか1からなることを特徴とするフラーレンの製 造装置。

【請求項3】 請求項1又は2記載のフラーレンの製造 装置において、前記各外側冷却部には水又は水以外の冷 媒のいずれか1を流通させることを特徴とするフラーレ ンの製造装置。

【語求項4】 語求項1~3のいずれか1項に記載のフラーレンの製造装置において、前記反応炉の内部に、前 20 記燃燒生成ガス流を分割する仕切壁が設けられ しかも 該仕切壁の内側には冷却能力が独立して制御可能な複数の内側冷却部を前記燃燒生成ガス流の流れ方向に対して多段に設けたことを特徴とするフラーレンの製造装置。

【請求項5】 請求項4記載のフラーレンの製造装置において、前記各内側冷却部が冷却管又は冷却ジャケットのいずれか1からなることを特徴とするフラーレンの製造装置。

【語求項6】 語求項4又は5記載のフラーレンの製造 30 装置において、前記各内側冷却部には、ガス、水、水以 外の冷媒のいずれか!を流通させることを特徴とするフ ラーレンの製造装置。

# 【発明の詳細な説明】

#### [0001]

【発明の届する技術分野】本発明は、フラーレンの製造 装置に関する。

# [0002]

【従来の技術】フラーレンは、ダイヤモンド、黒鉛に次 ぐ第三の炭素同素体の総称であり、Csc、Csc等に代表 40 されるように5員環と6員環のネットワークで閉じた中 空設状の炭素分子である。フラーレンの存在が最終的に 確認されたのは比較的最近の1990年のことであり、 比較的新しい炭素材料であるが、その特殊な分子構造ゆ えに特異的な物理的性質を示すことが認められ。例えば 以下のような広範囲な分野に渡り、革新的な用途開発が 急速に帰聞されつつある。

(1) 超硬材料への応用: フラーレンを前駆体とするこ ンとなる点で製造効率は良くない。しかし、副製する煤とで微細結晶粒子をもつ人工ダイヤモンドの製造が可能 を液体燃料等に使用可能なこと、製造装置が単純であるなため、付加価値のある耐摩耗材料への利用が期待され 50 こと等の点で、アーク台成法に対抗するフラーレンの大

#### ている。

(2) 医薬品への応用: C。誘導体、光デバイスを用いることで抗癌剤、エイズ・骨粗鬆症・アルツハイマー治療薬、造影剤、ステント材料等の用途としての研究が進められている。

(3) 超伝導材料への応用: フラーレン薄膜に金属カリウムをドープすると18Kという高い転移温度を持つ超伝導材料をつくり出すことができることが発見され、多方面から注目を集めている。

10 (4) 半導体製造への応用:レジストにC。。を混ぜることでレジスト構造がより一層強化されることを利用し、 次世代半導体製造への応用が期待されている。

【0003】 各種炭素数のフラーレンの中でもC。。及びC。。は比較的合成が容易であり、それゆえ今後の需要も爆発的に高まるととが予想されている。現在知られているフラーレンの製造方法としては以下に示す方法が挙げられる。

#### (1)レーザ蒸着法

希ガス中に置かれた炭素ターゲットに高エネルギー密度 のパルスレーザを照射し、炭素原子を蒸発させてフラー レンを合成する方法。電気炉内に不活性ガスが流れる石 英管を貫通させ、グラファイト試料をその石英管の中に 置く。ガスの流れの上流側からグラファイト試料にレー ザを照射し炭素原子を蒸発させると、電気炉出口付近の 石英管の内壁にC。。やC。。などのフラーレンを含む煤が 付着する。しかし、パルスレーザの1ショット当たりの 蒸発量がわずかであり、フラーレンの大畳製造には不向 きである。

#### (2)抵抗加熱法

50 大気圧以下のヘリウムガス雰囲気中でグラファイト棒を 通電加熱し、炭素原子を昇華させる方法。グラファイト 棒で構成した通電回路での電気抵抗ロスが大きいので、 フラーレンの大量製造には不向きである。

## (3) アーク放電法

数十k P a に保持したヘリウムガス雰囲気中で2本のグラファイト電極を軽く接触させたり あるいは1~2 m 加程度離した状態でアーク放電を起こし、陽極側の炭素原子を昇等させる方法。現在、工場規模でのフラーレンの大量製造に用いられている。

## 19 (4)高周波誘導加熱法

高周波誘導により原料グラファイトに過電液を流し、発生するシュール熱により原料グラファイトを加熱して炭素原子を蒸発する方法。

#### (5) 燃焼法

へりウム等の不活性ガスと酸素との混合ガス中でベンゼン等の炭化水素原料を不完全燃焼させる方法。ベンゼン 燃料の数%が製となり、その内の10%程度がフラーレンとなる点で製造効率は良くない。しかし、副製する煤 を液体燃料等に使用可能なこと、製造装置が単純であること等の点で、アーカ会球等に対抗するフラーレンの大 置生産法として注目されている。

#### (6) ナフタレン熱分解法

ナフタレンを約1000℃で熱分解させる方法。

【0004】以上のように、現在までにさまざまなフラ ーレンの合成法が提案されているが、いずれの方法にお いても、これまでにフラーレンを安価に大量に製造する 方法は確立されていない。これらの方法のうち、最も安 価で、効率的な製造方法の一つと考えられるのは燃烧法 であり、特表平6-507879号公報には、炭素含有 物を火炎中で燃焼させ経織物を収集することによるフラ 16 ーレンの製造方法が記載されている。この製造方法は、 炭素含有物を火炎中で燃漉させることによりフラーレン を製造するもので、実質的に燃焼のための燃料とフラー レンの原料は同一の炭素含有物となっている。フラーレ ンはすず状物質中に含まれて生成されるが、このすず状 物質の一部はいわゆるカーボンブラックである。 カーボ ンブラックの製造方法としては、ファーネス法、チャン ネル法、サーマル法、アセチレン法などが知られてお り、工業的な一般的製造方法としてはファーネス法が学 げられる。この方法では、例えば、円筒状の反応炉を使 20 用し、第1反応帯域で炉軸に対して水平方向又は垂直方 向に空気などの酸素含有ガスと燃料を供給して燃焼ガス 流を生じさせ、得られた燃焼ガス流を炉輪方向の下流に 設置され第1反応帯域と比較して縮小された断面積を有 する第2反応帯域に移動させ、そこで燃焼ガス流中に炭 素質原料、例えば炭化水素(原料油)を供給し反応させ てカーボンブラックを生成させる。次いで、カーボンブ ラックを含有した燃焼ガス流を、第2反応帯域の下流に ある第3反応帯域に移動させ、燃焼ガス流に冷却水の噴 霧などの冷却処理を行って燃焼ガスを急冷して反応を停 30 止させ、カーボンブラックを回収している。

#### [0005]

【発明が解決しようとする課題】しかしながら、上記の 通常のカーボンブラックの製造方法では、フラーレンは ほとんど生成しない。従って、フラーレンの製造におい ては、得られるすず状物質中に含まれるフラーレンの含 有割合をいかに高めるかが、大きな課題となっている。 フラーレンの収率を向上させるためには火炎温度を上昇 させる必要があることが特表平6-507879号公報 に記載されており、その手段として外部エネルギー源か ち火炎に更にエネルギーを供給する方法が述べられてい る。そして、好ましいエネルギー額としては、入力流の 電気抵抗加熱。マイクロウェーブ加熱、放電加熱及び向 流加熱が挙げられている。火炎温度を上げることによ り、炭素質原料の熱分解が促進されると共に高温の燃焼 生成ガスが得られて、フラーレン前駆体の形成には有利 となる。しかし、火炎温度の上昇は、燃焼生成ガス中に 形成されたフラーレン前駆体の分解を促進することにも なる。このため、形成されたフラーレン前駆体が燃焼生 成ガス中に安定して滞在できる存在時間が短くなって、 フラーレンの収率が低下する。更に、生成したフラーレンを回収するためにはフラーレンを含んだ燃焼生成ガスの温度を回収に最適な温度まで下げる必要がある。このため、燃焼生成ガスの温度が高過ぎると冷却装置の増強や冷却時間が長くなり、フラーレンの製造コストを増加させる要因となる。

【0006】フラーレンは次世代を担う新材料。新素材として多方面から注目されており、とのような燃煙生成ガスの高温化による炭素質原料の熱分解促進、燃煙生成ガスの温度の最適化によるフラーレン前駆体の形成と存在時間の確保。及び生成したフラーレンを含んだ燃焼生成ガスの冷却によるフラーレン回収の効率化を過不足なく全て達成することが可能なフラーレンの製造において、フラーレンの反応炉内の温度分布を制御することによりフラーレン前駆体の形成と安定化を確保してフラーレンを大量。安価、容易に製造して回収することが可能なフラーレンの製造装置を提供することを目的とする。

#### [0007]

【課題を解決するための手段】本発明者らは、燃焼法に よるフラーレンの大量かつ安価な製造方法におけるフラ ーレンの生成過程を穏々検討した結果。 フラーレンの反 応炉内の温度分布を制御することで、燃焼生成ガス中で のフラーレン前駆体の形成と安定化を促進して、フラー レン前駆体を効率的にフラーレンに成長させることがで きることを見出し、本発明を完成させた。前記目的に沿 う本発明に係るフラーレンの製造装置は、炭素含有化合 物供給口と酸素含有ガス供給口とを有する燃焼用バーナ 部を構える反応炉で、原料となる炭素含有化合物と酸素 含有ガスとを燃焼させてフラーレンを製造するフラーレ ンの製造装置であって、前記反応炉の外側には、冷却能 力を独立して制御可能な複数の外側冷却部を、前記炭素 含有化合物が前記酸素含有ガスの下で燃焼して形成され る燃燒生成ガス流の移動方向に対して多段に設けた。 【0008】反応炉の外側に、冷却能力を独立して制御 することが可能な外側冷却部を反応炉内の燃焼生成ガス

することが可能な外側冷却部を反応炉内の燃焼生成ガスの移動方向に対して多段に設けることによって、各外側冷却部と対応した反応炉の内側の温度を、燃焼生成ガスの移動方向に対してそれぞれ任意に訓御することが可能となる。その結果、反応炉の内側の温度に基づいて、燃焼生成ガスの温度を変化させることができる。例えば、炭素含有化合物の熱分解が促進される領域では高温の燃焼生成ガスが得られるように、反応炉の内側温度を管理する。その結果、反応活性の高い熱分解生成物を燃焼生成ガスのに存在させることができる。また、フラーレン前駆体が最初に形成される領域では、燃焼生成ガスの温度が保持されるように反応炉の内側温度を管理する。その結果、フラーレン前駆体の形成を促進することができる。更に、フラーレン前駆体が燃焼生成ガス流と共に移

動しながら成長してフラーレンになる領域では、形成さ れたフラーレン前駆体が安定して存在し成長できるよう に、反応炉の内側温度を管理する。その結果、フラーレ ン前駆体の移動と共に国囲の燃焼生成ガスの温度を徐々 に低下させることができ、フラーレン前駆体からフラー レンを効率的に生成させることができる。次いで、フラ ーレンの生成が完了した領域では、燃焼生成ガスの温度 を更に低下させることができるように、反応炉の内側温 度を管理する。その結果、フラーレンを含んだ燃煙生成 ガスの温度を、フラーレンの回収に最適な温度とするこ 19 度を制御することができる。 とができる。

【①①09】本発明に係るフラーレンの製造装置におい て、前記各外側冷却部が冷却管又は冷却ジャケットのい ずれかりからなる構成とすることができる。冷却管を使 用することにより簡便、安価に外側冷却部を形成するこ とができる。その際、冷却管の取付け間隔、冷却管の 径、冷却管に流す液体の種類と液置等を調整することに より冷却能力を変化させることができる。また、水冷ジ ャケットで外側冷却部を形成することにより、外側冷却 ジャケット内を流れる流体の流路、冷却ジャケットの容 荷、冷却ジャケットに流す流体の種類と流置等を調整す ることにより冷却能力を変化させることができる。更 に、冷却管で形成された外側冷却部と、冷却ジャケット で形成された外側冷却部を併用することにより、部位毎 に広範囲の冷却能力の調整を行うことができる。すなわ ち、大きな冷却能力を必要とする部位には冷却ジャケッ トを設置し、冷却能力をあまり必要としない部位には冷 却管を設置して、最適な冷却を行うことができる。

て、前記各外側冷却部には水又は水以外の冷媒のいずれ かしを流通させることができる。外側冷却部に水を流通 させることにより、安価にかつ簡便に冷却を行うことが できる。また、外側冷却部に、水以外の冷媒、例えば熱 模論等のように沸点が異なる冷模を使用することによ り、冷却能力を広範囲に調整することができる。更に、 水を流通させた外側冷却部と水以外の冷媒を流通させた 外側冷却部を併用することにより、各外部冷却部の部位 毎に冷却能力を広範囲に調整することができる。

【①①11】本発明に係るフラーレンの製造装置におい 40 て、前記反応炉の内部に、前記燃焼生成ガス流を分割す る仕切壁を設け、しかも、該仕切壁の内側には冷却能力 が独立して制御可能な複数の内側冷却部を前記燃度生成 ガス流の流れ方向に対して多段に設けた構成とすること ができる。反応炉内に燃焼生成ガス流を分割する仕切壁 を設け、この仕切壁の内側に冷却能力を独立して副御す ることが可能な内側冷却部を設けることによって、各内 側冷却部に対応した仕切壁の炉内側の表面温度を燃焼生 成ガスの移動方向に対してそれぞれ任意に制御すること

基づいて、焼焼生成ガスの温度を変化させることができ る。そして、炭素含有化合物の熱分解が促進される領域 では高温の燃焼生成ガスが得られるように、フラーレン 前駆体が最初に形成される領域では燃焼生成ガスの温度 が保持されるように、また。フラーレン前駆体が燃焼生 成ガス流と共に移動しながら成長してフラーレンになる 領域では形成されたフラーレン前駆体が安定して存在し 成長できるように、更に、プラーレンの生成が完了した。

領域では燃焼生成ガスの温度を更に低下させるように温

【りり12】本発明に係るフラーレンの製造装置におい て、前記各内側冷却部が冷却管又は冷却ジャケットのい ずれかしからなる構成とすることができる。冷却管を使 用することにより簡便、安価に内側冷却部を形成するこ とができる。その際、冷却管の取付け間隔、冷却管の 径、冷却質に流す液体の種類と流置等を調整することに より冷却能力を変化させることができる。また、水冷ジ ャケットで内側冷却部を形成することにより、内側冷却 部の冷却能力を向上させることができる。その際、冷却 部の冷却能力を向上させることができる。その際、冷却 20 ジャケット内を流れる液体の流路、冷却ジャケットの容 荷、冷却ジャケットに流す流体の種類と流畳等を調整す ることにより冷却能力を変化させることができる。更 に、冷却管と冷却ジャケットを併用することにより、部 位毎における冷却能力を広範囲に調整することができ る。すなわち、大きな冷却能力を必要とする部位には冷 却ジャケットを設置し、冷却能力をあまり必要としない 部位には冷却管を設置して、最適な冷却を行うことがで

【0013】本発明に係るフラーレンの製造装置におい 【0010】本発明に係るフラーレンの製造装置におい。30 て、前記各内側冷却部には、ガス、水、水以外の冷媒の いずれかしを流通させることができる。内側冷却部に水 を流道させることにより、安価にかつ簡便に冷却を行う ことができる。内側冷却部に、ガス、水以外の冷媒、例 えば熱媒抽等のように沸点が異なる冷媒を使用すること により、冷却能力を広範囲に調整することができる。ガ ス、水、水以外の冷媒を流通させた内側冷却部を併用す るととにより、各内側冷却部の各部位毎に冷却能力を広 範囲に調整することができる。

#### $\{0014\}$

【発明の実施の形態】続いて、添付した図面を参照しつ つ、本発明を具体化した実施の形態につき説明し、本発 明の理解に供する。ことに、図1(A)、(B)はそれ ぞれ本発明の第1の実施の形態に係るフラーレンの製造 装置の全体概略断面図、炭素含有化合物と酸素含有ガス の供給口の配置を示した燃焼用バーナ部の説明図、図2 (A)、(B)はそれぞれ本発明の第2の実施の形態に 係るフラーレンの製造装置の全体機略断面図、住切壁の 平面図である。図1(A)に示すように、本発明の第1 の実施の形態に係るフラーレンの製造装置 10は、反応 が可能となる。その結果。住切壁の炉内側の表面温度に 50 炉11と、反応炉11の下部に設けられた燃焼用バーナ

部12とを有している。以下、これらについて詳細に説 明する。反応炉11は、例えば、円筒形状の側壁部13 と、側壁部13の一端側に接続して徐々に外径が縮小し て排出口14を形成している端部壁15とを備えてい る。側壁部13と蟾部壁15は、例えばステンレス鋼等 の耐熱師で構成されている。 更に、側壁部13の他端側 の内周面には耐火物16がライニングされている。耐火 物16としては、例えばアルミナ質の耐火熄耳やアルミ ナ質の不定形耐火物を使用することができる。また、排 出口14には排気管17の一端側が接続され、他端側は 19 図示しない排気ポンプに接続されている。このため、反 応炉11内を大気圧未満の減圧状態にすると共に、反応 炉11内で生成したフラーレンを含む燃焼生成ガスを反 応炉11内から排気管17を介して外部に排出すること ができる。

【0015】側壁部13の一端側及び他端側の外層側に は冷却ジャケットからなる外側冷却部18、19.20 が、また外側冷却部18及び外側冷却部19の間には冷 却管からなる外側冷却部21が設けられている。また、 冷却部22が設けられている。更に、排気管17の外周 には冷却管からなる冷却部23が取付けられている。各 外側冷却部18.19、20、22には、例えば、流入 口aから水を流入させて流出口りから排出させることが でき、流通させる水の温度、流量、圧力を調整すること により各外側冷却部18.19、20.22の冷却能力 を調整できる。外側冷却部21及び冷却部23には、例 えば、流入口でから水を流入させて流出口でから排出さ せることができ、冷却ジャケットの場合と同様に流通さ せる水の温度、流量、圧力を調整することにより外側冷 30 却部21と冷却部23の冷却能力を調整できる。なお、 外側冷却部21を構成する冷却管の巻き間隔は、上方に 向かうにつれて徐々に小さくなるように設定されてい る。このため、外側冷却部21は、外側冷却部18側か **5外側冷却部19側に向かうにつれて冷却能力が徐々に** 向上する構造となっている。

【0016】以上のように、各外側冷却部18.19、 20.21、22を燃焼生成ガスの移動方向に対して多 段に設けることにより、燃焼生成ガスの移動方向。すな わち燃焼用バーナ部12側から排出口14側に向けて、 反応炉11の内側温度を徐々に低下させることがが可能 となる。従って、炭素含有化合物の燃焼と熱分解が生じ る領域では、発生した熱が奪われ難くなって、高温の燃 焼生成ガスが得られるようになる。また、フラーレン前 駆体が最初に形成される領域でも、燃焼生成ガスの温度 が保持されるようになる。更に、フラーレン前駆体が然 焼生成ガス流と共に移動しながら成長してフラーレンに なる領域では、フラーレン前駆体の移動と共に周囲の燃 焼生成ガスの温度を徐々に低下させるように制御すると は、燃焼生成ガスの温度を更に低下させることができ る.

【0017】側壁部13の他堤側に取付けられた燃焼用 バーナ部12は、例えばステンレス鋼等の耐熱鋼で形成 された基盤24と、基盤24に設けられ炭素含有化合物 を吐出する複数の供給口(炭素含有化合物供給口)2 5.26と、酸素含有ガスを吐出する複数の供給口(酸 素含有ガス供給口)27.28とを備えている。図1 (B) に示すように、炭素含有化合物の供給口25、2 6と、酸素含有ガスの供給口27、28は、各々独立に 分散して基盤24に設けられている。そして、各供給口 25.26、27、28の一方側は反応炉11の内側に 関口し、他方側は反応炉11の外側に開口していずれも 図示しない炭素含有化合物の供給管、酸素含有ガスの供 給管にそれぞれ接続されている。なお、炭素含有化合物 の供給管には加熱器が設けられており、反応炉11の内 部に吐出される際の炭素含有化合物の温度を、例えば2 0.0℃まで加温することができる。反応炉1.1の内側に 関口している各供給口25、26、27、28の形状は 鑑部壁15の外周側には、冷却ジャケットからなる外側 20 任意であり、略円形、楕円状、三角形や四角形などの多 角形状、ひょうたん型などの不定形状であってもよい。 本発明者らの知見によれば、円形よりも、長円径や長方 形のように長径と短径を持つ形状の方が、酸素含有ガス の加熱や希釈の速度がより速まる。従って、炭素含有化 合物の供給口25、26としては、精円状や略円形が好 ましく、酸素含有ガスの供給口27、28としては、ス リット状などの長方形状が好ましく、これらを組み合わ せるのが特に好ましい。

【0018】炭素含有化合物の供給口25、26と、酸 素含有ガスの供給口27、28の配置は、各々独立に反 応炉11の内側に関口していれば任意とできる。炭素含 有化合物の種類や、各供給口25、26、27,28の 数などのフラーレンの製造装置10の設計条件に合わせ ていろいろな配置を採用することができるが、例えば、 各々の供給口をフラーレンの製造装置10の軸心に対し て同心円周上に、周方向に交互に配置するならば、反応 炉11内での燃焼状態がより均一となるので好ましい。 この際に、酸素含有ガスの供給口の形状が長径及び短径 を持つような場合には、長径から延びた直線が円の中心 を通るように配置するのが好ましい。また、反応炉11 の内側に関口しているいずれの供給口25、26.2 7. 286、その関口蟾部が基盤24の表面と実質的に 同一平面上にあっても、突出していてもよいが、好まし くは実質的に同一平面上がよい。炭素含有化合物の供給 □25、26及び酸素含有ガスの供給□27、28から 反応炉11内に供給される炭素含有化合物及び酸素含有 ガスの各流は、 基々の供給 125、26、27、28の **端部から反応炉11内に対して任意の角度で供給してよ** いが、好ましくは基盤24に対して実質的に垂直となる とができる。更に、フラーレンの生成が完了する領域で 50 ように、夏には、供給される炭素含有化合物及び/又は 酸素含有ガスが供給口25.26、27、28の開口線 部の中心から実質的に同心円状に拡散するように供給するのが好ましい。

【10019】次に、本発明の第1の実施の形態に係るフ ラーレンの製造装置 1 ()を適用したフラーレンの製造方 法について詳細に説明する。炭素含有化合物の供給口2 5.26から供給する炭素含有化合物の畳と酸素含有ガ スの供給口27、28から供給する酸素ガス量を調整し て炭素含有化合物が不完全燃焼する条件で反応炉11内 に供給すると共に、俳気管17に接続された図示しない。19 鎌気ポンプで反応炉11内を大気圧未満、より好ましく は10~300 torrの状態に保持して、図示しない 着火手段で炭素含有化合物の燃焼を開始する。ここで、 炭素含有化合物と酸素含有ガスは各々独立し距離を隔て 分散配置されたそれぞれ複数の供給口25、26.2 7.28から反応炉11内に吐出されるため、反応炉1 1内における燃焼状態を均一にすることができる。ま た、炭素含有化合物の温度を、例えば、200℃に高め ることにより、炭素含有化合物が均一に燃焼することを 助長できる。更に、酸素含有ガス中の酸素ガス濃度はアー29 一様な流れが主体となる。 ルゴンガス等の不活性ガスにより希釈されて低下してい るととに加えて、反応炉11内の圧力が大気圧未満とな っているため、反応炉11内での繁焼状態を高温空気繁 焼状態と類似した状態にすることができる。その結果、 炭素含有化合物の燃焼が均一に進行して、反応炉11内 の温度を均一かつ高温(例えば、1000~1900 ℃. 好ましくは1700~1900℃) にすることがで

【0020】酸素含有ガスとしては、酸素源である酸素 ガスにアルゴンガス等の不活性ガスを任意の割合で混合 したガス(例えば、不活性ガスの濃度を()、又は()を超 えて90モル%以下の範囲で調整できる)を使用するこ とができる。酸素源としては、フラーレンの収率という 観点からは酸素ガスが好ましく、酸素源の入手のし易さ 等の額点からは空気が好ましい。特に燃焼温度を上げる ため、これらの酸素含有ガスは炉内に供給される前に予 熱することが好ましい。予熱の方法としては、熱交換器 を使用した禁煙生成ガスとの熱交換。 いわゆるリジェネ レーションバーナ等、公知のいかなる方法を用いても良 い。この予熱の温度は鴬温以上であればいかなる温度で も良いが、フラーレンの収率をあげるためには極力高温 度の方が好ましい。より好ましくは、炭素含有化合物の 自己着火温度以上であることが好ましい。炭素含有化合 物としては、一酸化炭素、天然ガス、石油ガス等の燃料 ガス、重独などの石油系液体燃料、クレオソート油など の石炭系液体燃料を使用することができる。中でもこれ ちを精製した芳香族系炭化水素を用いることが好まし く、特にベンゼンやトルエン等の芳香族系炭化水素が好 ましい。炭素含有化合物の純度は高い方が好ましく、中

近いほど好ましい。またフラーレンの収率を上げるため には、炭素含有化合物も不活性ガス等を用いて参釈する ことが好ましい。

【10021】続いて、炭素含有化合物が酸素含有ガスの 下で燃焼及び熱分解して形成される燃焼生成ガス流の温 度変化について説明する。炭素含有化合物の焼焼により 反応炉11内での燃焼領域の温度は、例えば、1000 ~1900℃、好ましくは1700~1900℃の高温 になる。このため、未燃焼の炭素含有化合物は容易に熱 分解して気化して、炭素含有化合物の燃焼により発生し た燃焼ガス内に拡散して燃焼生成ガスを形成する。反応 炉11内の圧力が大気圧未満で酸素ガス濃度が低い希薄 状態での燃焼であること、更に、均一燃焼が促進されて 燃燒生成ガスの温度が反応炉11の軸方向に垂直な方向 では実質的に一様になっていることから、燃焼生成ガス により形成される燃焼生成ガス流内では自己循環流が発 生しにくくなっている。そして、蒸焼生成ガスは排出口 14から排気ポンプで排気されているので、蒸煙生成ガ ス流は蒸焼用バーナ部12側から排出口14側に向かう

【0022】反応炉11の側壁部13と端部壁15の外 周側に設けられた外部冷却部18、19、20、21、 22の各冷却能力は、反応炉11の燃焼用バーナ部12 側から排出口14側に向かって徐々に大きくなるように 調整されている。このため、外側冷却部18、19、2 0.21、22と対向した反応炉11の内面温度を、燃 焼生成ガスの移動方向に対し徐々に低下させることが可 能となる。このため、反応炉11内を燃焼用バーナ部1 2個から排出口14個に向かって移動する燃焼生成ガス は、移動しながら反応炉11の内面温度に基づいて冷却 されることになる。従って、炭素含有化合物の熱分解が 促進される領域では冷却能力を低くしているため、高温 の燃焼生成ガスが得られる。その結果、反応活性の高い 熱分解生成物を燃焼生成ガス中に存在させることができ る。また、フラーレン前駆体が最初に形成される領域で も冷却能力が低いため、燃焼生成ガスの温度が保持され る。その結果、フラーレン前駆体の形成が促進される。 更に、フラーレン前躯体が燃焼生成ガス流と共に移動し ながら成長してフラーレンになる領域では、冷却能力が 徐々に大きくなるので、フラーレン前駆体の移動と共に 周囲の蒸焼生成ガスの温度が徐々に低下する。その結 果、フラーレン前駆体の分解が抑制されてフラーレンを 効率的に生成させることができる。更に、フラーレンの 生成が完了する領域では、燃焼生成ガスの温度を更に低 下させることができる。その結果、フラーレンを含んだ 燃煙生成ガスの温度を、フラーレンの回収に最適な温度 とすることができる。

く、特にベンゼンやトルエン等の芳香族系炭化水素が好 【① 023】図2(A)に示すように、本発明の第2のましい。炭素含有化合物の純度は高い方が好ましく、中 実施の形態に係るフラーレンの製造装置29は、反応炉でも芳香族系炭化水素を用いる際には純度が100%に 50 30と、反応炉30の下部に設けられた燃焼用バーナ部

31と、反応炉30内に設けられ燃焼生成ガス流を分割 する仕切壁32とを有している。以下、これらについて 詳細に説明するが、第1の実施の形態と実質的に同一の 構成要素については同一の符号を付して詳細な説明は省 略する。

【0024】仕切壁32は、図2(B)に示すよろに排 気管17内を挿道したステンレス鋼等の耐熱鋼で構成さ れた支持部材33と、一端側が支持部材33の側面に取 付けられ他端側が反応炉30の内面に当接して燃焼生成 ガス流を分割(当実施の形態では4分割)する4枚の分 10 るいは冷却ジャケットのみを設けることも可能である。 割壁34とを有している。 各分割壁34の内側には、支 持部村33の長手方向に、4段に積層され、それぞれ冷 却ジャケットからなる内側冷却部35、36、37、3 8が設けられている。各内側冷却部35、36.37、 38には、例えば、図示しない流入口から水を流入させ て図示しない流出口から排出させることができ、流通さ せる水の温度、流置、圧力を調整することにより各内側 冷却部35、36、37.38の冷却能力を独立して調 整できる。その結果、仕切壁32を構成する分割壁34 下させることが可能となる。

【0025】次に、本発明の第2の実施の形態に係るフ ラーレンの製造装置29を適用したフラーレンの製造方 法について説明する。 燃焼用バーナ部31から反応炉3 ①内に供給する炭素含有化合物と酸素含有ガスの構成及 び供給条件は、第1の実施の形態の場合と同様に設定す ることができるので、炭素含有化合物が酸素含有ガスの 下で燃焼及び熱分解して燃焼生成ガス流を形成する過程 及び燃焼生成ガス流内でフラーレンが生成する過程は、 第1の実施の形態で説明した場合と同様となる。従っ て、仕切壁32を設けたことによる燃焼生成ガス流の温 度変化について説明する。反応炉30の外側に設けた外 側冷却部18.19、20.21、22の冷却能力と、 仕切壁32に設けた内側冷却部35,36、37,38 の冷却能力をそれぞれ調整することにより、燃焼生成ガ スの移動方向に対して、反応炉30の側壁部13の内面 の温度分布、及び仕切壁32の表面の温度分布を実質的 に一致させることができる。従って、反応炉30内を燃 焼用バーナ部31側から排出口14側に向かって移動す る燃煙生成ガスに対して、側壁部13と仕切壁32は実 40 質的に同一の冷却を行うことができる。その結果、側壁 部13側の燃焼生成ガスも、仕切壁32側の燃焼生成ガ スも同じように冷却されて、反応炉30内部の燃煙生成 ガスの温度を水平方向では実質的に一様にすることがで きる。このため、反応炉30の内径が大きくなっても、 燃燒用バーナ部31側から排出口14側に向けて移動す る燃煙生成ガスの温度を、燃焼生成ガスの移動方向に対 して厳密に制御することができる。

【10026】以上、本発明の実施の形態を説明したが、 本発明は、この実施の形態に限定されるものではなく、 12

例えば、仕切壁を耐熱銅製の分割壁で構成したが、耐熱 銅製の分割壁の表面に耐火物をライニングしてもよい。 耐火物をライニングすることにより、仕切壁の耐熱性を 向上させることができ、反応炉内の更に高温領域から燃 焼生成ガスの温度を制御することができる。仕切壁に設 けた各内側冷却部を全て冷却ジャケットとしたが、全 て、冷却管とすることもできる、更に、冷却管と冷却ジ ャケットを併用することもできる。 外側冷却部として冷 却ジャケットと冷却管を併用したが、冷却管でのみ、あ 外側冷却部及び内側冷却部にはいずれも水を流通させた が、全ての外側冷却部に水以外の冷媒を流通させたり、 あるいは、水及び水以外の冷媒を流通させた外側冷却部 を併用することもできる。また、全ての内側冷却部にガ ス又は水以外の冷媒を流通させたり、あるいは、ガス、 水、水以外の冷媒から任意に選んだ2以上の冷媒を流通 させることもできる。

[0027]

【発明の効果】請求項1~6記載のフラーレンの製造装 の表面温度を燃燒生成ガスの移動方向に対して徐々に低 20 置においては、反応炉の外側には、冷却能力を独立して 制御可能な複数の外側冷却部を、炭素含有化合物が酸素 含有ガスの下で燃焼して形成される燃焼生成ガス流の移 動方向に対して多段に設けたので、反応炉内の燃焼生成 ガスの流れ方向に対する温度分布を副御することによ り、燃焼生成ガスの高温化による炭素含有化合物の熱分 解促進、燃焼生成ガス中におけるフラーレン前駆体の形 成促進、フラーレン前駆体の存在時間の確保、及び生成 したフラーレンを含んだ燃焼生成ガスの冷却によるフラ ーレン回収の効率化をそれぞれ図ることが可能となる。 30 その結果、フラーレンを大量、容易に製造して回収する ことが可能となってフラーレンの製造コストを低減させ ることが可能となる。

> 【0028】特に、請求項2記載のフラーレンの製造装 置においては、各外側冷却部が冷却管又は冷却ジャケッ トのいずれか1からなるので、簡便、安価に反応炉の外 壁内側湿度を副御して燃煙生成ガスの移動方向に対する 温度分布を制御することが可能となる。その結果。フラ ーレンの生成率の向上及びフラーレン回収の効率化を図 ることが可能となる。また、冷却管と冷却ジャケットか らなる外側冷却部を併用することで冷却能力の調整を行 うことができ、反応炉内での燃焼生成ガスの移動方向に 対する温度分布を厳密に制御することが可能となる。そ の結果、フラーレンの生成率の向上及びフラーレン回収 の効率化を更に図ることが可能となる。

【0029】請求項3記載のフラーレンの製造装置にお いては、各外側冷却部には水又は水以外の冷媒のいずれ か」を流通させるので、水又は水以外の冷媒を目的に合 わせて選択することで冷却能力を変えて冷却することに より、燃焼生成ガスの流れ方向に対する燃焼生成ガス流 50 の温度分布を効果的に制御することが可能となる。その 結果、フラーレンの生成率の向上及びフラーレン回収の 効率化を図ることが可能となる。また、水と水以外の冷 媒を併用することで冷却能力の調整を行うことができ、 反応炉内での燃焼生成ガスの移動方向に対する温度分布 を厳密に制御することが可能となる。その結果、フラー レンの生成率の向上及びフラーレン回収の効率化を更に 図ることが可能となる。

【0030】語求項4記載のフラーレンの製造装置にお いては、反応炉の内部に、燃焼生成ガス歳を分割する仕 切壁が設けられ、しかも、仕切壁の内側には冷却能力が 19 独立して制御可能な複数の内側冷却部を蒸焼生成ガス流 の流れ方向に対して多段に設けたので、反応炉内での燃 焼生成ガスの移動方向に対する温度分布を、反応炉内の 外側と中央部側で一様となるように副御することが可能 となる。その結果、大型の反応炉においても、反応炉内 での燃焼生成ガスの移動方向に対する温度分布を厳密に 制御することができ、フラーレンを大量に製造して回収 することが可能となって、フラーレンの製造コストを更 に低減させることが可能となる。

【0031】請求項5記載のフラーレンの製造装置にお 20 仕切壁の平面図である。 いては、各内側冷却部が冷却管又は冷却ジャケットのい ずれか1からなるので、簡優、安価に反応炉内に設けた 仕切壁の表面温度を制御して燃焼生成ガスの移動方向に 対する温度分布を制御することが可能となる。その結 果、フラーレンの生成率の向上及びフラーレン回収の効 率化を図ることが可能となる。また、冷却管と冷却ジャ ケットからなる内側冷却部を併用することで冷却能力の 調整を行うことができ、反応炉内での燃焼生成ガスの移 動方向に対する温度分布を厳密に制御することが可能と なる。その結果、フラーレンの生成率の向上及びフラー 36 7 38 内側冷却部

レン回収の効率化を更に図ることが可能となる。

【0032】請求項6記載のフラーレンの製造装置にお いては、各内側冷却部には、ガス、水、水以外の冷媒の いずれかしを流道させるので、それぞれ最適な冷却能力 を遵択して蒸焼生成ガスの流れ方向に対する反応炉内の 温度分布を効果的に制御することが可能になる。更に、 反応炉の外側と中央部側で温度が一様となるように制御 することが可能となる。その結果、大型の反応炉におい ても、燃焼生成ガスの流れ方向に対する反応炉内の温度 分布を効果的に副御することができ、フラーレンを大量 に製造して回収することが可能となって、フラーレンの 製造コストを更に低減させることが可能となる。

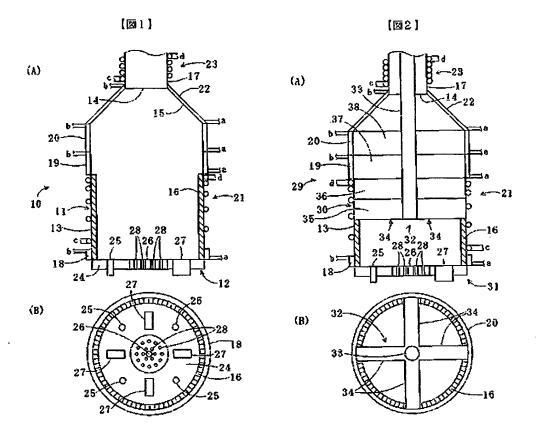
## 【図面の簡単な説明】

【図1】(A). (B) はそれぞれ本発明の第1の実施 の影態に係るフラーレンの製造装置の全体機関断面図、 炭素含有化合物と酸素含有ガスの供給口の配置を示した 燃焼用バーナ部の説明図である。

【図2】(A). (B)はそれぞれ本発明の第2の実施 の形態に係るフラーレンの製造装置の全体機略断面図、

### 【符号の説明】

10:フラーレンの製造装置、11:反応炉、12:燃 焼用バーナ部、13:側壁部、14:排出口、15:端 部壁. 16:耐火物、17:排気管. 18、19. 2 0.21、22:外側冷却部、23:冷却管、24:基 盤. 25、26:炭素含有化合物の供給口、27.2 8:酸素含有ガスの供給口、29:フラーレンの製造装 置、30:反応炉、31:燃焼用バーナ部、32:仕切 壁. 33:支持部材、34:分割壁. 35、36.3



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## CLAIMS

# [Claim(s)]

[Claim 1] With a fission reactor equipped with the burner section for combustion which has a carbon content compound feed hopper and oxygen content gas supply opening It is the manufacturing installation of the fullerene which the carbon content compound and oxygen content gas used as a raw material are burned, and manufactures fullerene. In the outside of said fission reactor The manufacturing installation of the fullerene characterized by preparing refrigeration capacity independently in multistage to the migration direction of a combustion generation gas stream in which said carbon content compound is formed by burning under said oxygen content gas in two or more controllable outside cooling sections.

[Claim 2] The manufacturing installation of the fullerene characterized by said each outside cooling section consisting of any 1 of a cooling pipe or the cooling jackets in the manufacturing installation of fullerene according to claim 1.

[Claim 3] The manufacturing installation of the fullerene characterized by circulating any 1 of the refrigerants other than water or water in said each outside cooling section in the manufacturing installation of fullerene according to claim 1 or 2.

[Claim 4] The manufacturing installation of the fullerene characterized by having established the bridge wall which divides said combustion generation gas stream into the interior of said fission reactor, and moreover refrigeration capacity preparing independently two or more controllable inside cooling sections in multistage to the flow direction of said combustion generation gas stream in the manufacturing installation of fullerene given in any 1 term of claims 1-3 inside this bridge wall. [Claim 5] The manufacturing installation of the fullerene characterized by said each inside cooling section consisting of any 1 of a cooling pipe or the cooling jackets in the manufacturing installation of fullerene according to claim 4.

[Claim 6] The manufacturing installation of the fullerene characterized by circulating any 1 of the refrigerants other than gas, water, and water in the manufacturing installation of fullerene according to claim 4 or 5 at said each inside cooling section.

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## **DETAILED DESCRIPTION**

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacturing installation of fullerene. [0002]

[Description of the Prior Art] Fullerene is the generic name of the third carbon allotrope which ranks second to a diamond and a graphite, and is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings so that it might be represented by C60 and C70 grade. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient: since manufacture of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs: research as an application of an anticancer agent, an acquired immunode-ficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material: if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture: it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.
- [0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.
- (1) How to irradiate the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, evaporate a carbon atom, and compound fullerene. Into an electric furnace, the quartz tube with which inert gas flows is made to penetrate, and a graphite sample is placed into the quartz tube. If laser is irradiated from the upstream of the flow of gas at a graphite sample and a carbon atom is evaporated, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube near an electric furnace outlet. However, the evaporation per shot of a pulse laser is slight, and it is unsuitable for extensive manufacture of fullerene.
- (2) The approach to which carry out energization heating of the graphite rod in the gaseous helium ambient atmosphere below resistance heating method atmospheric pressure, and a carbon atom is made to sublimate. Since the electric resistance loss in the energization circuit constituted from a graphite rod is large, it is unsuitable for extensive manufacture of fullerene.
- (3) the-ten number of arc discharge methods -- the approach to which the carbon atom by the side of a

lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium ambient atmosphere held to kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in current and a works scale.

(4) How to heat an eddy current to raw material graphite by radio frequency heating method high frequency induction, heat raw material graphite with a sink and the Joule's heat to generate, and evaporate a carbon atom.

- (5) The approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. Manufacture effectiveness is not good at the point that several% of a benzene fuel serves as soot, and about 10% of them becomes fullerene. However, the soot which sub\*\* is observed as a method of mass-producing the fullerene which opposes an arc synthesis method in respect of a manufacturing installation being usable to liquid fuel etc., and simple etc.
- (6) The approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0004] As mentioned above, although the synthesis method of various fullerene by current is proposed. in which approach, the method of manufacturing fullerene in large quantities cheaply until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [six to] official report, and collecting condensates in it is indicated. When this manufacture approach burns a carbon inclusion in a flame, fullerene is manufactured and the fuel for combustion and the raw material of fullerene serve as the same carbon inclusion substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black. As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is mentioned as the industrial general manufacture approach. By this approach, use a cylinderlike fission reactor, supply oxygen content gas and fuels, such as air, to horizontal or a perpendicular direction to \*\*\*\* in the 1st reaction band, for example, and a combustion gas style is produced. Make it move to the 2nd reaction band which has the cross section which was installed in the lower stream of a river of furnace shaft orientations in the obtained combustion gas style, and was reduced as compared with the 1st reaction band, and supply a carbonaceous raw material (stock oil), for example, a hydrocarbon, it is made then, to react into a combustion gas style, and carbon black is made to generate. Subsequently, the combustion gas style containing carbon black is moved to the 3rd reaction band on the lower stream of a river of the 2nd reaction band, cooling processing of spraying of cooling water etc. is performed in the style of combustion gas, combustion gas is quenched, a reaction is stopped, and carbon black is collected.

[0005]

[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. Therefore, in manufacture of fullerene, it has been a big technical problem how the content rate of the fullerene contained in the soot-like matter obtained is raised. In order to raise the yield of fullerene, it is indicated by the Patent Publication Heisei No. 507879 [six to] official report that it is necessary to raise flame temperature, and the approach of supplying energy to a flame further from an external energy source as the means is described. And as a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned. By raising flame temperature, while the pyrolysis of a carbonaceous raw material is promoted, hot combustion generation gas is obtained, and it becomes advantageous to formation of a fullerene precursor. However, the rise of flame temperature is also promoting disassembly of the fullerene precursor formed into combustion generation gas. For this reason, the existence time amount at which the formed fullerene precursor is stabilized in combustion generation gas, and can stay becomes short, and the yield of fullerene falls. Furthermore, in order to collect the generated fullerene, it is necessary to lower the temperature of the combustion generation gas containing fullerene to the optimal temperature for recovery. For this reason, if the temperature of

combustion generation gas is too high, enhancement and the cooldown delay of a cooling system will become long, and will become the factor which makes the manufacturing cost of fullerene increase. [0006] Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the manufacturing installation of fullerene possible in attaining all the neither more nor less in the increase in efficiency of the fullerene recovery by formation of the fullerene precursor by pyrolysis promotion of the carbonaceous raw material by elevated-temperature-izing of such combustion generation gas and optimization of the temperature of combustion generation gas and cooling of the combustion generation gas containing the fullerene which existence time amount secured and generated is desired. This invention was made in view of this situation, and aims at securing formation and stabilization of a fullerene precursor and offering the manufacturing installation of extensive and the fullerene which it manufactures cheaply and easily and can be collected for fullerene by controlling the temperature distribution in the fission reactor of fullerene in manufacture of the fullerene by the combustion method.

[0007]

[Means for Solving the Problem] this invention persons completed a header and this invention for the ability of a fullerene precursor for formation and stabilization of a fullerene precursor in combustion generation gas to be promoted, and to be efficiently grown up into fullerene by controlling the temperature distribution in the fission reactor of fullerene, as a result of examining various generation processes of the fullerene in the extensive and cheap manufacture approach of fullerene by the combustion method. The manufacturing installation of the fullerene concerning this invention in alignment with said purpose With a fission reactor equipped with the burner section for combustion which has a carbon content compound feed hopper and oxygen content gas supply opening It is the manufacturing installation of the fullerene which the carbon content compound and oxygen content gas used as a raw material are burned, and manufactures fullerene. In the outside of said fission reactor Refrigeration capacity was prepared independently in multistage to the migration direction of a combustion generation gas stream in which said carbon content compound is formed by burning under said oxygen content gas in two or more controllable outside cooling sections.

[0008] It becomes possible to control the temperature inside each outside cooling section and the corresponding fission reactor to arbitration to the direction of a combustion generation firedamp migration, respectively by preparing the outside cooling section which can control refrigeration capacity independently in the outside of a fission reactor to the direction of a combustion generation firedamp migration in a fission reactor multistage. Consequently, the temperature of combustion generation gas can be changed based on the temperature inside a fission reactor. For example, in the field in which the pyrolysis of a carbon content compound is promoted, the inside temperature of a fission reactor is managed so that hot combustion generation gas may be obtained. Consequently, the high pyrolysate of labile can be made to exist in combustion generation gas. Moreover, in the field in which a fullerene precursor is formed first, the inside temperature of a fission reactor is managed so that the temperature of combustion generation gas may be held. Consequently, formation of a fullerene precursor can be promoted. Furthermore, in the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene, the inside temperature of a fission reactor is managed so that the formed fullerene precursor may be stabilized and it can exist and grow up. Consequently, the temperature of surrounding combustion generation gas can be gradually reduced with migration of a fullerene precursor, and fullerene can be made to generate efficiently from a fullerene precursor. Subsequently, in the field which generation of fullerene completed, the inside temperature of a fission reactor is managed so that the temperature of combustion generation gas can be reduced further. Consequently, temperature of the combustion generation gas containing fullerene can be made into the optimal temperature for recovery of fullerene.

[0009] In the manufacturing installation of the fullerene concerning this invention, it can consider as the configuration which said each outside cooling section becomes from any 1 of a cooling pipe or the cooling jackets. The outside cooling section can be formed simple and cheaply by using a cooling pipe. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which are passed

to anchoring spacing of a cooling pipe, the path of a cooling pipe, and a cooling pipe in that case. Moreover, the refrigeration capacity of the outside cooling section can be raised by forming the outside cooling section with a water cooled jacket. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which pass the inside of a cooling jacket to the passage of flowing fluid, the volume of a cooling jacket, and a cooling jacket in that case. Furthermore, wide range refrigeration capacity can be adjusted for every part by using together the outside cooling section formed with the cooling pipe, and the outside cooling section formed with the cooling jacket. That is, a cooling jacket can be installed in the part which needs big refrigeration capacity, a cooling pipe can be installed in the part which seldom needs refrigeration capacity, and optimal cooling can be performed. [0010] In the manufacturing installation of the fullerene concerning this invention, any 1 of the refrigerants other than water or water can be circulated in said each outside cooling section. By circulating water in the outside cooling section, it can cool cheaply and simple. Moreover, refrigeration capacity can be broadly adjusted by using refrigerants other than water, for example, the refrigerant with which the boiling points differ like a thermal oil, for the outside cooling section. Furthermore, refrigeration capacity can be broadly adjusted for every part of each external-intercooling section by using together the outside cooling section which circulated water, and the outside cooling section which circulated refrigerants other than water.

[0011] In the manufacturing installation of the fullerene concerning this invention, the bridge wall which divides said combustion generation gas stream can be prepared in the interior of said fission reactor, and, moreover, it can consider as the configuration in which refrigeration capacity prepared independently two or more controllable inside cooling sections in multistage to the flow direction of said combustion generation gas stream inside this bridge wall. It becomes possible to control the skin temperature inside [ furnace ] the bridge wall corresponding to each inside cooling section to arbitration to the direction of a combustion generation firedamp migration, respectively by establishing the bridge wall which divides a combustion generation gas stream in a fission reactor, and preparing the inside cooling section which can control refrigeration capacity independently inside this bridge wall. Consequently, the temperature of combustion generation gas can be changed based on the skin temperature inside [furnace] a bridge wall. And so that hot combustion generation gas may be obtained in the field in which the pyrolysis of a carbon content compound is promoted So that the temperature of combustion generation gas may be held in the field in which a fullerene precursor is formed first Moreover, so that the fullerene precursor formed in the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene may be stabilized and it can exist and grow up Furthermore, temperature is controllable by the field which generation of fullerene completed to reduce the temperature of combustion generation gas further.

[0012] In the manufacturing installation of the fullerene concerning this invention, it can consider as the configuration which said each inside cooling section becomes from any 1 of a cooling pipe or the cooling jackets. The inside cooling section can be formed simple and cheaply by using a cooling pipe. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which are passed to anchoring spacing of a cooling pipe, the path of a cooling pipe, and a cooling pipe in that case. Moreover, the refrigeration capacity of the inside cooling section can be raised by forming the inside cooling section with a water cooled jacket. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which pass the inside of a cooling jacket to the passage of flowing fluid, the volume of a cooling jacket, and a cooling jacket in that case. Furthermore, the refrigeration capacity in every part can be broadly adjusted by using a cooling pipe and a cooling jacket together. That is, a cooling jacket can be installed in the part which needs big refrigeration capacity, a cooling pipe can be installed in the part which seldom needs refrigeration capacity, and optimal cooling can be performed. [0013] In the manufacturing installation of the fullerene concerning this invention, any 1 of the refrigerants other than gas, water, and water can be circulated in said each inside cooling section. By circulating water in the inside cooling section, it can cool cheaply and simple. Refrigeration capacity can be broadly adjusted by using refrigerants other than gas and water, for example, the refrigerant with which the boiling points differ like a thermal oil, for the inside cooling section. Refrigeration capacity

can be broadly adjusted for every each part grade of each inside cooling section by using together the inside cooling section which circulated refrigerants other than gas, water, and water.
[0014]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented.

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# **TECHNICAL FIELD**

[Field of the Invention] This invention relates to the manufacturing installation of fullerene.

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## PRIOR ART

[Description of the Prior Art] Fullerene is the generic name of the third carbon allotrope which ranks second to a diamond and a graphite, and is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings so that it might be represented by C60 and C70 grade. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient: since manufacture of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs: research as an application of an anticancer agent, an acquired immunode-ficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material: if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture: it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.
- [0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below as the manufacture approach of fullerene learned now is mentioned.
- (1) How to irradiate the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, evaporate a carbon atom, and compound fullerene. Into an electric furnace, the quartz tube with which inert gas flows is made to penetrate, and a graphite sample is placed into the quartz tube. If laser is irradiated from the upstream of the flow of gas at a graphite sample and a carbon atom is evaporated, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube near an electric furnace outlet. However, the evaporation per shot of a pulse laser is slight, and it is unsuitable for extensive manufacture of fullerene.
- (2) The approach to which carry out energization heating of the graphite rod in the gaseous helium ambient atmosphere below resistance heating method atmospheric pressure, and a carbon atom is made to sublimate. Since the electric resistance loss in the energization circuit constituted from a graphite rod is large, it is unsuitable for extensive manufacture of fullerene.
- (3) the-ten number of arc discharge methods the approach to which the carbon atom by the side of a lifting and an anode plate is made to sublimate arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium ambient atmosphere held to kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in current and a works scale.

State of the State

(4) How to heat an eddy current to raw material graphite by radio frequency heating method high

frequency induction, heat raw material graphite with a sink and the Joule's heat to generate, and evaporate a carbon atom.

- (5) The approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. Manufacture effectiveness is not good at the point that several% of a benzene fuel serves as soot, and about 10% of them becomes fullerene. However, the soot which sub\*\* is observed as a method of mass-producing the fullerene which opposes an arc synthesis method in respect of a manufacturing installation being usable to liquid fuel etc., and simple etc.
- (6) The approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0004] As mentioned above, although the synthesis method of various fullerene by current is proposed, in which approach, the method of manufacturing fullerene in large quantities cheaply until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, and the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the Patent Publication Heisei No. 507879 [six to] official report, and collecting condensates in it is indicated. When this manufacture approach burns a carbon inclusion in a flame, fullerene is manufactured and the fuel for combustion and the raw material of fullerene serve as the same carbon inclusion substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black. As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is mentioned as the industrial general manufacture approach. By this approach, use a cylinderlike fission reactor, supply oxygen content gas and fuels, such as air, to horizontal or a perpendicular direction to \*\*\*\* in the 1st reaction band, for example, and a combustion gas style is produced. Make it move to the 2nd reaction band which has the cross section which was installed in the lower stream of a river of furnace shaft orientations in the obtained combustion gas style, and was reduced as compared with the 1st reaction band, and supply a carbonaceous raw material (stock oil), for example, a hydrocarbon, it is made then, to react into a combustion gas style, and carbon black is made to generate. Subsequently, the combustion gas style containing carbon black is moved to the 3rd reaction band on the lower stream of a river of the 2nd reaction band, cooling processing of spraying of cooling water etc. is performed in the style of combustion gas, combustion gas is quenched, a reaction is stopped, and carbon black is collected.

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#### EFFECT OF THE INVENTION

[Effect of the Invention] In the manufacturing installation of fullerene according to claim 1 to 6 In the outside of a fission reactor, since refrigeration capacity was prepared in multistage to the migration direction of a combustion generation gas stream in which a carbon content compound is formed independently by burning under oxygen content gas, two or more controllable outside cooling sections. By controlling the temperature distribution over the flow direction of the combustion generation gas in a fission reactor Pyrolysis promotion of the carbon content compound by elevated-temperature-izing of combustion generation gas, the promotion of formation of the fullerene precursor in combustion generation gas, It becomes possible to attain the increase in efficiency of the fullerene recovery by cooling of the combustion generation gas containing the fullerene which fullerene precursor's existence time amount secured and generated, respectively. Consequently, it becomes possible to become possible to manufacture fullerene in large quantities and easily and to collect it, and to reduce the manufacturing cost of fullerene.

[0028] Especially, since each outside cooling section consists of any 1 of a cooling pipe or the cooling jackets in the manufacturing installation of fullerene according to claim 2, it becomes possible to control the outer wall inside temperature of a fission reactor simple and cheaply, and to control the temperature distribution over the direction of a combustion generation firedamp migration. Consequently, it becomes possible to attain the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery. Moreover, refrigeration capacity can be adjusted by using together the outside cooling section which consists of a cooling pipe and a cooling jacket, and it becomes possible to control strictly the temperature distribution over the direction of a combustion generation firedamp migration in a fission reactor. Consequently, it becomes possible to attain further the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery.

[0029] In the manufacturing installation of fullerene according to claim 3, since any 1 of the refrigerants other than water or water is circulated in each outside cooling section, it becomes possible by changing refrigeration capacity and cooling by choosing [ for the purpose of refrigerants other than water or water ], to control effectively the temperature distribution of the combustion generation gas stream over the flow direction of combustion generation gas. Consequently, it becomes possible to attain the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery. Moreover, refrigeration capacity can be adjusted by using together refrigerants other than water and water, and it becomes possible to control strictly the temperature distribution over the direction of a combustion generation firedamp migration in a fission reactor. Consequently, it becomes possible to attain further the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery. [0030] In the manufacturing installation of fullerene according to claim 4, since the bridge wall which divides a combustion generation gas stream was prepared in the interior of a fission reactor and refrigeration capacity moreover prepared independently two or more controllable inside cooling sections in multistage to the flow direction of a combustion generation gas stream inside the bridge wall, it becomes possible to control the temperature distribution over the direction of a combustion generation firedamp migration in a fission reactor to become uniform at the outside in a fission reactor, and a

center-section side. Consequently, also in a large-sized fission reactor, the temperature distribution over the direction of a combustion generation firedamp migration in a fission reactor can be controlled strictly, it becomes possible to manufacture fullerene in large quantities and to collect it, and it becomes possible to reduce the manufacturing cost of fullerene further.

[0031] In the manufacturing installation of fullerene according to claim 5, since each inside cooling section consists of any 1 of a cooling pipe or the cooling jackets, it becomes possible to control the skin temperature of the bridge wall established in the fission reactor simple and cheaply, and to control the temperature distribution over the direction of a combustion generation firedamp migration. Consequently, it becomes possible to attain the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery. Moreover, refrigeration capacity can be adjusted by using together the inside cooling section which consists of a cooling pipe and a cooling jacket, and it becomes possible to control strictly the temperature distribution over the direction of a combustion generation firedamp migration in a fission reactor. Consequently, it becomes possible to attain further the improvement in the yield of fullerene, and the increase in efficiency of fullerene recovery.

[0032] In the manufacturing installation of fullerene according to claim 6, since any 1 of the refrigerants other than gas, water, and water is circulated, it becomes possible to choose the respectively optimal refrigeration capacity and to control effectively the temperature distribution in the fission reactor to the flow direction of combustion generation gas at each inside cooling section. Furthermore, it becomes possible to control so that temperature becomes uniform by the outside of a fission reactor, and the center-section side. Consequently, also in a large-sized fission reactor, the temperature distribution in the fission reactor to the flow direction of combustion generation gas can be controlled effectively, it becomes possible to manufacture fullerene in large quantities and to collect it, and it becomes possible to reduce the manufacturing cost of fullerene further.

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#### TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the abovementioned usual carbon black, fullerene is hardly generated. Therefore, in manufacture of fullerene, it has been a big technical problem how the content rate of the fullerene contained in the soot-like matter obtained is raised. In order to raise the yield of fullerene, it is indicated by the Patent Publication Heisei No. 507879 [ six to ] official report that it is necessary to raise flame temperature, and the approach of supplying energy to a flame further from an external energy source as the means is described. And as a desirable energy source, electric resistance heating of an input style, microwave heating, discharge heating, and counterflow heating are mentioned. By raising flame temperature, while the pyrolysis of a carbonaceous raw material is promoted, hot combustion generation gas is obtained, and it becomes advantageous to formation of a fullerene precursor. However, the rise of flame temperature is also promoting disassembly of the fullerene precursor formed into combustion generation gas. For this reason, the existence time amount at which the formed fullerene precursor is stabilized in combustion generation gas, and can stay becomes short, and the yield of fullerene falls. Furthermore, in order to collect the generated fullerene, it is necessary to lower the temperature of the combustion generation gas containing fullerene to the optimal temperature for recovery. For this reason, if the temperature of combustion generation gas is too high, enhancement and the cooldown delay of a cooling system will become long, and will become the factor which makes the manufacturing cost of fullerene increase. [0006] Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the manufacturing installation of fullerene possible in attaining all the neither more nor less in the increase in efficiency of the fullerene recovery by formation of the fullerene precursor by pyrolysis promotion of the carbonaceous raw material by elevatedtemperature-izing of such combustion generation gas and optimization of the temperature of combustion generation gas and cooling of the combustion generation gas containing the fullerene which existence time amount secured and generated is desired. This invention was made in view of this situation, and aims at securing formation and stabilization of a fullerene precursor and offering the manufacturing installation of extensive and the fullerene which it manufactures cheaply and easily and can be collected for fullerene by controlling the temperature distribution in the fission reactor of fullerene in manufacture of the fullerene by the combustion method.

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## **MEANS**

[Means for Solving the Problem] this invention persons completed a header and this invention for the ability of a fullerene precursor for formation and stabilization of a fullerene precursor in combustion generation gas to be promoted, and to be efficiently grown up into fullerene by controlling the temperature distribution in the fission reactor of fullerene, as a result of examining various generation processes of the fullerene in the extensive and cheap manufacture approach of fullerene by the combustion method. The manufacturing installation of the fullerene concerning this invention in alignment with said purpose With a fission reactor equipped with the burner section for combustion which has a carbon content compound feed hopper and oxygen content gas supply opening It is the manufacturing installation of the fullerene which the carbon content compound and oxygen content gas used as a raw material are burned, and manufactures fullerene. In the outside of said fission reactor Refrigeration capacity was prepared independently in multistage to the migration direction of a combustion generation gas stream in which said carbon content compound is formed by burning under said oxygen content gas in two or more controllable outside cooling sections. [0008] It becomes possible to control the temperature inside each outside cooling section and the corresponding fission reactor to arbitration to the direction of a combustion generation firedamp migration, respectively by preparing the outside cooling section which can control refrigeration capacity independently in the outside of a fission reactor to the direction of a combustion generation firedamp migration in a fission reactor multistage. Consequently, the temperature of combustion generation gas can be changed based on the temperature inside a fission reactor. For example, in the field in which the pyrolysis of a carbon content compound is promoted, the inside temperature of a fission reactor is managed so that hot combustion generation gas may be obtained. Consequently, the high pyrolysate of labile can be made to exist in combustion generation gas. Moreover, in the field in which a fullerene precursor is formed first, the inside temperature of a fission reactor is managed so that the temperature of combustion generation gas may be held. Consequently, formation of a fullerene precursor can be promoted. Furthermore, in the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene, the inside temperature of a fission reactor is managed so that the formed fullerene precursor may be stabilized and it can exist and grow up. Consequently, the temperature of surrounding combustion generation gas can be gradually reduced with migration of a fullerene precursor, and fullerene can be made to generate efficiently from a fullerene precursor. Subsequently, in the field which generation of fullerene completed, the inside temperature of a fission reactor is managed so that the temperature of combustion generation gas can be reduced further. Consequently, temperature of the combustion generation gas containing fullerene can be made into the optimal temperature for recovery of fullerene.

[0009] In the manufacturing installation of the fullerene concerning this invention, it can consider as the configuration which said each outside cooling section becomes from any 1 of a cooling pipe or the cooling jackets. The outside cooling section can be formed simple and cheaply by using a cooling pipe. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which are passed to anchoring spacing of a cooling pipe, the path of a cooling pipe, and a cooling pipe in that case.

Moreover, the refrigeration capacity of the outside cooling section can be raised by forming the outside cooling section with a water cooled jacket. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which pass the inside of a cooling jacket to the passage of flowing fluid, the volume of a cooling jacket, and a cooling jacket in that case. Furthermore, wide range refrigeration capacity can be adjusted for every part by using together the outside cooling section formed with the cooling pipe, and the outside cooling section formed with the cooling jacket. That is, a cooling jacket can be installed in the part which needs big refrigeration capacity, a cooling pipe can be installed in the part which seldom needs refrigeration capacity, and optimal cooling can be performed. [0010] In the manufacturing installation of the fullerene concerning this invention, any 1 of the refrigerants other than water or water can be circulated in said each outside cooling section. By circulating water in the outside cooling section, it can cool cheaply and simple. Moreover, refrigeration capacity can be broadly adjusted by using refrigerants other than water, for example, the refrigerant with which the boiling points differ like a thermal oil, for the outside cooling section. Furthermore, refrigeration capacity can be broadly adjusted for every part of each external-intercooling section by using together the outside cooling section which circulated water, and the outside cooling section which circulated refrigerants other than water.

[0011] In the manufacturing installation of the fullerene concerning this invention, the bridge wall which divides said combustion generation gas stream can be prepared in the interior of said fission reactor, and, moreover, it can consider as the configuration in which refrigeration capacity prepared independently two or more controllable inside cooling sections in multistage to the flow direction of said combustion generation gas stream inside this bridge wall. It becomes possible to control the skin temperature inside [ furnace ] the bridge wall corresponding to each inside cooling section to arbitration to the direction of a combustion generation firedamp migration, respectively by establishing the bridge wall which divides a combustion generation gas stream in a fission reactor, and preparing the inside cooling section which can control refrigeration capacity independently inside this bridge wall. Consequently, the temperature of combustion generation gas can be changed based on the skin temperature inside [furnace] a bridge wall. And so that hot combustion generation gas may be obtained in the field in which the pyrolysis of a carbon content compound is promoted So that the temperature of combustion generation gas may be held in the field in which a fullerene precursor is formed first Moreover, so that the fullerene precursor formed in the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene may be stabilized and it can exist and grow up Furthermore, temperature is controllable by the field which generation of fullerene completed to reduce the temperature of combustion generation gas further.

[0012] In the manufacturing installation of the fullerene concerning this invention, it can consider as the configuration which said each inside cooling section becomes from any 1 of a cooling pipe or the cooling jackets. The inside cooling section can be formed simple and cheaply by using a cooling pipe. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which are passed to anchoring spacing of a cooling pipe, the path of a cooling pipe, and a cooling pipe in that case. Moreover, the refrigeration capacity of the inside cooling section can be raised by forming the inside cooling section with a water cooled jacket. Refrigeration capacity can be changed by adjusting a class, a flow rate, etc. of a fluid which pass the inside of a cooling jacket to the passage of flowing fluid, the volume of a cooling jacket, and a cooling jacket in that case. Furthermore, the refrigeration capacity in every part can be broadly adjusted by using a cooling pipe and a cooling jacket together. That is, a cooling jacket can be installed in the part which needs big refrigeration capacity, a cooling pipe can be installed in the part which seldom needs refrigeration capacity, and optimal cooling can be performed. [0013] In the manufacturing installation of the fullerene concerning this invention, any 1 of the refrigerants other than gas, water, and water can be circulated in said each inside cooling section. By circulating water in the inside cooling section, it can cool cheaply and simple. Refrigeration capacity can be broadly adjusted by using refrigerants other than gas and water, for example, the refrigerant with which the boiling points differ like a thermal oil, for the inside cooling section. Refrigeration capacity can be broadly adjusted for every each part grade of each inside cooling section by using together the

inside cooling section which circulated refrigerants other than gas, water, and water. [0014]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. They are the whole manufacturing installation outline sectional view of the fullerene which the whole manufacturing installation outline sectional view of the fullerene which drawing 1 (A) and (B) require for the gestalt of operation of the 1st of this invention, respectively, the explanatory view of the burner section for combustion having shown arrangement of the feed hopper of a carbon content compound and oxygen content gas, drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention here, respectively, and the top view of a bridge wall. As shown in drawing 1 (A), the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 1st of this invention has a fission reactor 11 and the burner section 12 for combustion prepared in the lower part of a fission reactor 11. Hereafter, these are explained to a detail. The fission reactor 11 is equipped with the cylindrical shape-like side-attachment-wall section 13 and the edge wall 15 which it connects with the end side of the side-attachment-wall section 13, and an outer diameter contracts gradually, and forms the exhaust port 14. The side-attachment-wall section 13 and the edge wall 15 consist of heat-resisting steel, such as stainless steel. Furthermore, refractories 16 are lined by the inner skin by the side of the other end of the side-attachment-wall section 13. As refractories 16, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of an exhaust pipe 17 is connected to an exhaust port 14, and the other end side is connected to the exhaust air pump which is not illustrated. For this reason, while changing the inside of a fission reactor 11 into the reduced pressure condition of under atmospheric pressure, the combustion generation gas containing the fullerene generated in the fission reactor 11 can be discharged outside through an exhaust pipe 17 from the inside of a fission reactor 11.

[0015] The outside cooling section 21 which the outside cooling sections 18, 19, and 20 which consist of a cooling jacket turn into from a cooling pipe between the outside cooling section 18 and the outside cooling section 19 again is formed in the periphery side by the side of the end of the side-attachmentwall section 13, and the other end. Moreover, the outside cooling section 22 which consists of a cooling jacket is formed in the periphery side of the edge wall 15. Furthermore, the cooling section 23 which consists of a cooling pipe is attached in the periphery of an exhaust pipe 17. Water can be made to be able to flow into each outside cooling sections 18, 19, 20, and 22 from Input a, they can be made to discharge from a tap hole b, and the refrigeration capacity of each outside cooling sections 18, 19, 20, and 22 can be adjusted to them by adjusting the temperature of the water to circulate, a flow rate, and a pressure. Water can be made to be able to flow into the outside cooling section 21 and the cooling section 23 from Input c, they can be made to discharge from a tap hole d, and the refrigeration capacity of the outside cooling section 21 and the cooling section 23 can be adjusted to them by adjusting the temperature of the water circulated like the case where it is a cooling jacket, a flow rate, and a pressure. In addition, volume spacing of the cooling pipe which constitutes the outside cooling section 21 is set up so that it may become small gradually, as it goes up. For this reason, the outside cooling section 21 has structure whose refrigeration capacity improves gradually as it goes to the outside cooling section 19 side from the outside cooling section 18 side.

[0016] As mentioned above, \*\* of reducing the inside temperature of a fission reactor 11 gradually becomes possible from a direction 12 of combustion generation firedamp migration, i.e., the burner section for combustion, side towards an exhaust port 14 side by forming each outside cooling sections 18, 19, 20, 21, and 22 in multistage to the direction of a combustion generation firedamp migration. Therefore, in the field which combustion and the pyrolysis of a carbon content compound produce, the generated heat becomes is hard to be taken, and hot combustion generation gas comes to be obtained. Moreover, the temperature of combustion generation gas comes to be held also in the field in which a fullerene precursor is formed first. Furthermore, it is controllable by the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene to reduce the temperature of surrounding combustion generation gas gradually with migration of a fullerene precursor.

Furthermore, in the field which generation of fullerene completes, the temperature of combustion generation gas can be reduced further.

[0017] The burner section 12 for combustion attached in the other end side of the side-attachment-wall section 13 is equipped with the base 24 formed with heat-resisting steel, such as stainless steel, two or more feed hoppers (carbon content compound feed hopper) 25 and 26 which are prepared in a base 24 and carry out the regurgitation of the carbon content compound, and two or more feed hoppers (oxygen content gas supply opening) 27 and 28 which carry out the regurgitation of the oxygen content gas. As shown in drawing 1 (B), it distributes independently respectively and the feed hoppers 25 and 26 of a carbon content compound and the feed hoppers 27 and 28 of oxygen content gas are formed in the base 24. And opening of the one side of each feed hoppers 25, 26, 27, and 28 is carried out inside a fission reactor 11, and the other side is connected to the supply pipe of the carbon content compound which is carrying out opening to the outside of a fission reactor 11, and does not illustrate a gap, either, and the supply pipe of oxygen content gas, respectively. In addition, the heater is formed in the supply pipe of a carbon content compound, and the temperature of the carbon content compound at the time of being breathed out inside a fission reactor 11 can be warmed to 200 degrees C. The configuration of each feed hoppers 25, 26, 27, and 28 which are carrying out opening inside the fission reactor 11 may be arbitrary. and may have the shape of an indeterminate form, such as the shape of a polygon, such as the shape of an approximate circle form and an ellipse, a triangle, and a square, and a gourd mold. According to this invention persons' knowledge, in the configuration in which a circular twist also has a major axis and a minor axis like the diameter of an ellipse, or a rectangle, heating of oxygen content gas and the rate of dilution speed up more. Therefore, as feed hoppers 25 and 26 of a carbon content compound, the shape of an ellipse and an approximate circle form are desirable, as feed hoppers 27 and 28 of oxygen content gas, the shape of a rectangle, such as the shape of a slit, is desirable, and it is desirable especially to combine these.

[0018] If opening of the arrangement of the feed hoppers 25 and 26 of a carbon content compound and the feed hoppers 27 and 28 of oxygen content gas is respectively carried out inside the fission reactor 11 independently, it will be possible with arbitration. Although various arrangement is employable according to the design condition of the manufacturing installation 10 of fullerene, such as a class of carbon content compound, and the number of each feed hoppers 25, 26, 27, and 28, if each feed hopper is arranged by turns on a concentric circle periphery to the axial center of the manufacturing installation 10 of fullerene in a hoop direction, since the combustion condition in a fission reactor 11 will become more uniform for example, it is desirable. In this case, when the configuration of the feed hopper of oxygen content gas has a major axis and a minor axis, it is desirable to arrange so that the straight line prolonged from the major axis may pass along the core of a circle. Moreover, although you may project even if any feed hoppers 25, 26, 27, and 28 which are carrying out opening inside the fission reactor 11 have the open end on the same flat surface substantially with the front face of a base 24, the same flatsurface top is substantially good preferably. Each \*\* of the carbon content compound supplied in a fission reactor 11 from the feed hoppers 25 and 26 of a carbon content compound and the feed hoppers 27 and 28 of oxygen content gas and oxygen content gas Although you may supply from the edge of each feed hoppers 25, 26, 27, and 28 at an angle of arbitration to the inside of a fission reactor 11, so that it may become perpendicular substantially to a base 24 preferably further It is desirable to supply so that the carbon content compound and/or oxygen content gas which are supplied may be substantially spread in concentric circular from the core of the open end of feed hoppers 25, 26, 27, and 28. [0019] Next, the manufacture approach of the fullerene which applied the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 1st of this invention is explained to a detail. While supplying in a fission reactor 11 on the conditions on which the amount of the carbon content compound supplied from the feed hoppers 25 and 26 of a carbon content compound and the amount of oxygen gas supplied from the feed hoppers 27 and 28 of oxygen content gas are adjusted, and a carbon content compound combusts incompletely The inside of a fission reactor 11 is more preferably held in the condition of 10 - 300torr under atmospheric pressure with the exhaust air pump which was connected to the exhaust pipe 17 and which is not illustrated, and combustion of a carbon content

compound is started with the ignition means which is not illustrated. Here, a carbon content compound and oxygen content gas become independent respectively, and since it is breathed out in a fission reactor 11 from the feed hoppers 25, 26, 27, and 28 of each plurality which separated distance and were distributed, they can make homogeneity the combustion condition in a fission reactor 11. Moreover, it can promote that a carbon content compound burns in homogeneity by raising the temperature of a carbon content compound to 200 degrees C. furthermore, the thing which the oxygen gas concentration in oxygen content gas is diluted by inert gas, such as argon gas, and is being fallen -- in addition, since the pressure in a fission reactor 11 has become under atmospheric pressure, the combustion condition in a fission reactor 11 can be changed into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of a carbon content compound advances to homogeneity, and can make temperature in a fission reactor 11 homogeneity and an elevated temperature (for example, 1000-1900 degrees C, preferably 1700-1900 degrees C). [0020] As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a furnace, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion generation gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature beyond , what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of a carbon content compound. As a carbon content compound, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, and creosote oil, can be used. Especially, it is desirable to use the aromatic series system hydrocarbon which refined these, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so desirable that its purity is close to 100% in case the purity of a carbon content compound uses an aromatic series system hydrocarbon especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute a carbon content compound using inert gas etc.

[0021] Then, the temperature change of the combustion generation gas stream in which it burns and pyrolyzes and a carbon content compound is formed under oxygen content gas is explained. 1000-1900 degrees C of temperature of the combustion zone in a fission reactor 11 become a 1700-1900-degree C elevated temperature preferably by combustion of a carbon content compound, for example. For this reason, a non-burned carbon content compound is pyrolyzed easily, is evaporated, is diffused in the combustion gas which occurred by combustion of a carbon content compound, and forms combustion generation gas. That the pressure in a fission reactor 11 is combustion in the thin condition that oxygen gas concentration is low, under in atmospheric pressure, and since homogeneity combustion is promoted and the temperature of combustion generation gas is uniform substantially in the direction perpendicular to the shaft orientations of a fission reactor 11 further, it has been hard coming to generate self-circulating flow within the combustion generation gas stream formed by combustion generation gas. And since combustion generation gas is exhausted with the exhaust air pump from the exhaust port 14, the uniform flow by which a combustion generation gas stream goes to an exhaust port 14 side from the burner section 12 side for combustion serves as a subject.

[0022] From the burner section 12 side for combustion of a fission reactor 11, each refrigeration capacity of the side-attachment-wall section 13 of a fission reactor 11 and the external-intercooling sections 18, 19, 20, 21, and 22 prepared in the periphery side of the edge wall 15 is adjusted so that it may become large gradually toward an exhaust port 14 side. For this reason, it becomes possible to reduce gradually the inside temperature of the outside cooling sections 18, 19, 20, 21, and 22 and the fission reactor 11 which countered to the direction of a combustion generation firedamp migration. For

this reason, while the combustion generation gas which moves toward an exhaust port 14 side from the burner section 12 side for combustion moves in the inside of a fission reactor 11, it will be cooled based on the inside temperature of a fission reactor 11. Therefore, in the field in which the pyrolysis of a carbon content compound is promoted, since refrigeration capacity is made low, hot combustion generation gas is obtained. Consequently, the high pyrolysate of labile can be made to exist in combustion generation gas. Moreover, since refrigeration capacity is low also in the field in which a fullerene precursor is formed first, the temperature of combustion generation gas is held. Consequently, formation of a fullerene precursor is promoted. Furthermore, in the field which grows while a fullerene precursor moves with a combustion generation gas stream, and becomes fullerene, since refrigeration capacity becomes large gradually, the temperature of surrounding combustion generation gas falls gradually with migration of a fullerene precursor. Consequently, disassembly of a fullerene precursor is controlled and fullerene can be made to generate efficiently. Furthermore, in the field which generation of fullerene completes, the temperature of combustion generation gas can be reduced further. Consequently, temperature of the combustion generation gas containing fullerene can be made into the optimal temperature for recovery of fullerene.

[0023] As shown in drawing 2 (A), the manufacturing installation 29 of the fullerene concerning the gestalt of operation of the 2nd of this invention has a fission reactor 30, the burner section 31 for combustion prepared in the lower part of a fission reactor 30, and the bridge wall 32 which is established in a fission reactor 30 and divides a combustion generation gas stream. Hereafter, although these are explained to a detail, the sign same about the same component is substantially attached with the gestalt of the 1st operation, and detailed explanation is omitted.

[0024] The bridge wall 32 has the supporter material 33 which consisted of heat-resisting steel, such as stainless steel which inserted in the inside of an exhaust pipe 17 as shown in drawing 2 (B), and the separating wall 34 of four sheets with which an end side is attached in the side face of the supporter material 33, and an other end side divides a combustion generation gas stream in contact with the inside of a fission reactor 30 (it quadrisects with the gestalt of this operation). Inside each separating wall 34, a laminating is carried out to four steps and the inside cooling sections 35, 36, 37, and 38 which consist of a cooling jacket, respectively are formed in the longitudinal direction of the supporter material 33. Each inside cooling sections 35, 36, 37, and 38 can be made to discharge from the tap hole which water is made to flow from the input which is not illustrated and is not illustrated, and the refrigeration capacity of each inside cooling sections 35, 36, 37, and 38 can be adjusted independently to them by adjusting the temperature of the water to circulate, a flow rate, and a pressure, for example. Consequently, it becomes possible to reduce gradually the skin temperature of the separating wall 34 which constitutes a bridge wall 32 to the direction of a combustion generation firedamp migration.

[0025] Next, the manufacture approach of the fullerene which applied the manufacturing installation 29 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained. Since the configuration and the conditions of supply of a carbon content compound and oxygen content gas which supply in a fission reactor 30 from the burner section 31 for combustion can set up like the case of the gestalt of the 1st operation, the process which fullerene generates within the process in which a carbon content compound burns and pyrolyzes under oxygen content gas, and forms a combustion generation gas stream, and a combustion generation gas stream becomes that it is the same as that of the case where the gestalt of the 1st operation explains. Therefore, the temperature change of the combustion generation gas stream by having established the bridge wall 32 is explained. The temperature distribution of the inside of the side-attachment-wall section 13 of a fission reactor 30 and the temperature distribution of the front face of a bridge wall 32 can be made substantially in agreement by adjusting the refrigeration capacity of the outside cooling sections 18, 19, 20, 21, and 22 prepared in the outside of a fission reactor 30, and the refrigeration capacity of the inside cooling sections 35, 36, 37, and 38 prepared in the bridge wall 32, respectively to the direction of a combustion generation firedamp migration. Therefore, the side-attachment-wall section 13 and a bridge wall 32 can perform the same cooling substantially to the combustion generation gas which moves toward an exhaust port 14 side in the inside of a fission reactor 30 from the burner section 31 side for combustion. Consequently, the combustion generation gas by the

side of a bridge wall 32 is cooled similarly, and if the combustion generation gas by the side of the side-attachment-wall section 13 is also horizontal, it can make uniform substantially temperature of the combustion generation gas of the fission reactor 30 interior. For this reason, even if the bore of a fission reactor 30 becomes large, the temperature of the combustion generation gas which moves towards an exhaust port 14 side from the burner section 31 side for combustion is strictly controllable to the direction of a combustion generation firedamp migration.

[0026] As mentioned above, although the gestalt of operation of this invention is explained, this invention is not limited to the gestalt of this operation, and although the bridge wall was constituted from a separating wall made from heat-resisting steel, refractories may be lined on the front face of the separating wall made from heat-resisting steel. By lining refractories, the thermal resistance of a bridge wall can be raised, it is in a fission reactor, and also the temperature of combustion generation gas can be controlled from an elevated-temperature field. Although each inside cooling sections of all prepared in the bridge wall were used as the cooling jacket, all can also be used as a cooling pipe, and also a cooling pipe and a cooling jacket can also be used together. Although the cooling jacket and the cooling pipe were used together as the outside cooling section, it is also possible to prepare only a cooling pipe or a cooling jacket. Although water was all circulated in the outside cooling section and the inside cooling section, refrigerants other than water can be circulated in all the outside cooling sections, or the outside cooling section which circulated refrigerants other than water and water can also be used together. Moreover, refrigerants other than gas or water can be circulated in all the inside cooling sections, or two or more refrigerants chosen as arbitration from refrigerants other than gas, water, and water can also be circulated.

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## **DESCRIPTION OF DRAWINGS**

# [Brief Description of the Drawings]

[Drawing 1] (A) and (B) are the whole manufacturing installation outline sectional view of the fullerene concerning the gestalt of operation of the 1st of this invention, and the explanatory view of the burner section for combustion having shown arrangement of the feed hopper of a carbon content compound and oxygen content gas, respectively.

[Drawing 2] (A) and (B) are the whole manufacturing installation outline sectional view of the fullerene concerning the gestalt of operation of the 2nd of this invention, and the top view of a bridge wall, respectively.

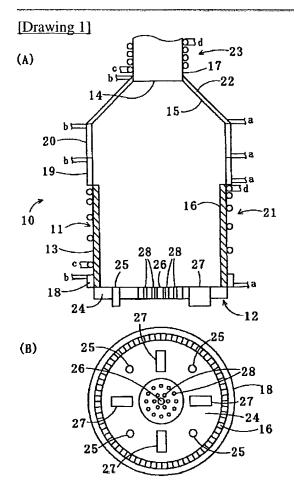
[Description of Notations]

The manufacturing installation of fullerene, 11:fission reactor, 12: 10: The burner section for combustion, 13: The side-attachment-wall section, 14:exhaust port, 15:edge wall, 16:refractories, 17: An exhaust pipe, 18, 19, 20, 21, 22:outside cooling section, 23:cooling pipe, 24: A base, The feed hopper of the feed hopper of 25 and 26:carbon content compound, 27, and 28:oxygen content gas, the manufacturing installation of 29:fullerene, 30:fission reactor, the burner section for 31:combustion, 32:bridge wall, 33:supporter material, 34:separating wall, 35, 36, 37, 38: Inside cooling section

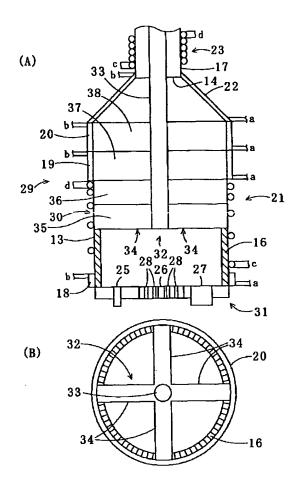
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# **DRAWINGS**



[Drawing 2]



[Translation done.]

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